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MAR. 11. 2004 2:01 PM0732 69143323 AGRO LINZ GMBH  
2027218250NR.396 S.2  
NO. 6108 P. 11

## IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re application of

: Confirmation No. 9813

Gerhard COUFAL

: Docket No. 2001-0462A

Serial No. 09/830,074

: Group Art Unit 1624

Filed April 23, 2001

: Examiner V. Balasubramanian

METHOD FOR PRODUCING PURE  
MELAMINEDECLARATION OF PETER WEISS UNDER 37 C.F.R. 1.132Commissioner for Patents  
P.O. Box 1450  
Alexandria, VA 22313-1450

Sir:

Peter Weiss declares that:

1. He is a citizen of Austria, residing at Steinweg 10, A-4061 Pasching, Austria;
2. His education and employment history are as follows:

In 1970 he received a M. Sc. (Dipl. Ing.) in from the Technical University, Vienna, Austria.

In 1974 he joined Chemie Linz, a predecessor of Agrolinz Melamin GmbH, where he worked in the Chemical Research Department (melamine field).

From 1983 to 1991 he was production assistant in the acrylonitrile plant in Enns and then, from 1992 to 1997 he was project manager for the translocation of the Enns acrylonitrile plant to South Africa.

From 1997 to 2002 he was head of the Melamin/Urea Production in Linz, where he was, in addition to the operation of 4 existing melamine plants, responsible for the start up of a new melamine production plant in the year 2000. This new plant is a wet cooling process similar to the present invention.

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During his work as head of melamine production he was also responsible for the melamine quality control laboratory.

Since 2003 he has been head of the melamine technology department where he still works very closely with the melamine production department. His responsibility in melamine technology is the continuous optimization of all the Agrolinz Melamine production plants, both at the Linz, Austria production site and the Castellanza, Italy melamine production site.

3. In the course of melamine process control, he conducted numerous experiments and interpreted numerous melamine analyses results and APHA measurements. For the quality control of the melamine he always used the HPLC technique with respect to by-product quantity and the APHA measurement with respect to the melamine control color evaluation. He knows that for a correct APHA measurement it is important to have low by-product content of melamine.

4. For Example 3 on page 8 of the response dated September 16, 2003 and the supplemental response of October 28, 2003 there was taken a sample directly after the quencher of the above mentioned new melamine plant in Linz. The analysis of the sample then was made by his quality control laboratory.

The example was run as follows:

#### 5. EXAMPLE 3

In a representative experiment of the present invention, the melamine was produced in high pressure melamine reactor at 373.5°C and 135 bar. The off gases were separated and the CO<sub>2</sub> was removed. Cooling of the melamine melt was effected by gaseous ammonia from 373.5°C (reactor temperature) to 361.5°C (temperature is 1 to 50°C above the melting point).

*Pete Werts* The resulting melt was then quenched with an aqueous ammonia solution. After the quenching, the melamine solution is in a solution. After the crystallization, the APHA value was 5.

6. Regarding Example 3, it is stated that the APHA value is obtained after crystallization.

Between quenching and crystallization, some additional process steps are performed. These process steps are not recited in main claim 15.

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To eliminate any possible issue as to whether the low APHA values are due to these process steps and not due to the invention as claimed, the following experiment was performed.

A sample from a melamine suspension was taken directly after quenching. The suspension was cooled to 20°C whereby the melamine crystal crystallized. The solid was washed out with a double amount of water. All other process conditions were identical to those in Example 3.

Due to some by-products, the melamine resin was slightly opaque. To facilitate the APHA measurement, some NaOH was added. It is important to note that the NaOH does not affect the APHA value itself and that the APHA value has nothing to do with the opacity caused by the by-products.

The measured APHA value directly after quenching was 35.

This shows that directly after quenching, i.e. only using process steps from the independent claim, the APHA value is smaller by at least a factor of three in comparison to the APHA values in Examples 1 and 2 which are discussed in the accompanying Declaration of Birgit Bogner as representative of Canzi, U.S. 5,721,313. This is a clear indication that the claimed invention has an unobvious advantage over Canzi. The positive effect on the APHA value could not be expected.

He further declares that all statements made herein of his own knowledge are true, and that all statements on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code, and that such willful false statements may jeopardize the validity of the application or any patent issuing thereon.

1.04.2004

Date



Peter WEISS

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## IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re application of

: Confirmation No. 9813

Gerhard COUFAL

: Docket No. 2001-0462A

Serial No. 09/830,074

: Group Art Unit 1624

Filed April 23, 2001

: Examiner V. Balasubramanian

METHOD FOR PRODUCING PURE  
MELAMINEDECLARATION NO. 1 OF BIRGIT BOGNER UNDER 37 C.F.R. 1.132Commissioner for Patents  
P.O. Box 1450  
Alexandria, VA 22313-1450

Sir:

Birgit Bogner declares that:

1. She is a citizen of Austria, residing at Albert Schöpf Straße 60, A-4020 Linz, Austria;
2. Her education and employment history are as follows:

In 1995 she received a M. Sc. (Dipl. -Ing.).

From April 1996 to June 1998 she worked in the Melamine Technology Department Of Agrolinz Melamin GmbH and was engaged in the development of a new high pressure production process for melamine. This research was done in a melamine pilot plant in Castellanza, Italy, where a subsidiary company of Agrolinz Melamin GmbH is situated.

Besides her work at the melamine pilot plant in Castellanza she was also responsible for the laboratory work concerning the pilot plant experiments.

In June 1998 she returned to Linz where a new melamine plant was built and where she assisted in the technological engineering and plant start up in 2000.

Since 2000 she has been patent referee of Agrolinz Melamin GmbH, where her special field is again melamine technology.

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3. During her 2 years work in the melamine pilot plant in Castellanza she conducted many experiments verifying the Canzi dry cooling process which is the subject of U.S. 5,721,363. Mr. Canzi, who developed the Castellanza pilot plant, was her predecessor in melamine technology in Agrolinz Melamine GmbH. For the evaluation of the pilot plant melamine they used the HPLC technique for the by-product quantity and the APHA measurement for the color quality. In performing the Canzi dry cooling method she always observed APHA values larger than 100.

4. Two of the numerous pilot plant experiments that she conducted in connection with the Canzi process were Examples 1 and 2 presented in the September 16, 2003 response to the Office Action dated May 16, 2003.

5. Examples 1 and 2 were representative of the Canzi process and were conducted as follows:

#### EXAMPLE 1

Molten melamine was kept at a pressure of 76 bar for 75 minutes. Subsequently, the melamine was cooled by a defined cooling program to 240°C, thereby solidifying the melamine. Then the melamine was depressurized and the melamine was slowly cooled to ambient temperature.

The analysis yielded the following results:

Ammelide	50 ppm
Ammeline	135 ppm
Ursidomelamine	50 ppm
Melam	410 ppm

The APHA value could not be determined due to the opacity of the liquid; the APHA value would have been excessively high.

#### EXAMPLE 2

Melamine was treated as above but is kept under a pressure of 105 bar for 90 minutes. The defined slow cooling was performed to 250°C.

The analysis yielded the following results:

Ammelide	50 ppm
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Ammeline 132 ppm

Ureidomelamine 50 ppm

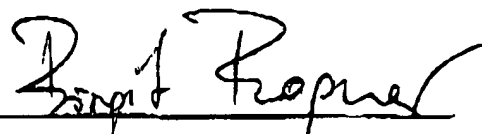
Melam 300 ppm

The APHA value could be measured, but was larger than 100;

She further declare that all statements made herein of her own knowledge are true, and that all statements on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code, and that such willful false statements may jeopardize the validity of the application or any patent issuing thereon.

25.03.2004

Date

  
Birgit BOGNER



IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re application of

Gerhard COUFAL

Serial No. 09/830,074

Filed April 23, 2001

Docket No. 2001-0462A

Group Art Unit 1624

Examiner V. Balasubramanian

METHOD FOR PRODUCING PURE  
MELAMINE

DECLARATION NO. 2 OF BIRGIT BOGNER UNDER 37 CFR 1.132

Birgit Bogner declares that:

1. She is a citizen of Austria, residing at Albert Schöpf Straße 60, A-4020 Linz, Austria;
2. Her education and employment history are as follows:

In 1995 she received a M. Sc. (Dipl. -Ing.).

From April 1996 to June 1998 she worked in the Melamine Technology Department of Agrolinz Melamin GmbH, engaged in the development of a new high pressure production process for melamine. This research was done in a melamine pilot plant in Castellanza, Italy, where a subsidiary company of Agrolinz Melamin GmbH is situated.

Besides her work at the melamine pilot plant in Castellanza she was also responsible for the laboratory work concerning the pilot plant experiments.

In June 1998 she returned to Linz where a new melamine plant was built and where she assisted in the technological engineering and plant start up in 2000.

Since 2000 she has been patent referee of Agrolinz Melamin GmbH, where her special field is again melamine technology.

3. The present invention is based on an advantageous method of recovering melamine from a melt prepared from urea in a high-pressure process. The instant method involves cooling of the melamine melt down to a temperature close to the solidification point i.e. melting point, but

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still above the melting point. In claim 15, this is expressed as "about 1 to 50°C above the melting point of melamine which is dependent on the ammonia pressure."

In the attached diagram, it can be seen how this first step is conducted, i.e. so that the molten melamine is cooled to a temperature just above the solidification point, by the addition of ammonia.

In the second cooling step, the melamine at just above the melting point is quenched with an aqueous solution such as water, aqueous ammonia etc., which is an inexpensive means for cooling.

This, the present invention employs water as a cooling agent to effect phase transition from the molten state to the solid state in an economical and efficient manner.

These features are neither disclosed nor suggested by the prior art as will be discussed in detail below.

Claims 15 to 17 and 19 to 21 have been rejected under 35 U.S.C. 102(b) as anticipated by Kokubo (U.S. 3,637,686) in the subject application.

In reply:

Kokubo employs a two step cooling process for recovering purified melamine which is very different from that presently claimed.

In a first step, crude molten melamine or a mixture of melamine with ammonia and carbon dioxide etc. passes through pipe 1 into a pressure cooler 2, in which the first stage i.e. cooling section 23 cools the molten melamine to a temperature range of 200-270°C to solidify it and in the second stage i.e. in cooling section 18, the product is cooled from 200-270°C to 100-200°C by quenching with aqueous ammonia. See column 4, lines 52-63.

Further, as pointed out in column 2, lines 30 et seq., the cooled molten melamine is cooled under pressure to 200-270°C (to avoid deammoniazation condensation and hydrolysis, which hardly occurs at less than 300°C) where it solidifies. After cooling to solidification of the melamine in the first step, the hydrolysis of the crude melamine can be well controlled by further cooling the solid melamine to 100-200°C by means of an aqueous solution containing ammonia (col. 2, lines 59 to 64).



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While Kokubo states that the crude molten (emphasis) melamine is cooled to 100-200°C by means of the aqueous solution containing ammonia, what is meant is that initially, the material being treated was molten, however, it became solidified at 200-270°C as pointed out in column 2, lines 58-60.

Therefore, the melamine cannot again be solidified by quenching into aqueous ammonia to bring it to a temperature of 100-200°C since it had been previously solidified at 200-270°C.

A comparison of the present process and Kokubo can be seen from the attached diagram in which the first step of the present process is conducted in the shaded area above the melting point line.

The second step is conducted in the present process below the melting point line.

On the other hand, in Kokubo, the first step involves solidification (of course below the melting point) and the aqueous quench takes place after the melamine has already been solidified in the preceding step.

There are several operational advantages to proceeding in this manner and these will be discussed below. However, even in the absence of these advantages, the steps themselves are simply unobvious from the teaching of Kokubo.

With regard to the advantages of the present method over Kokubo:

#### 1. Less Cooling Medium is Needed

In Kokubo, all of the cooling energy for the solidification has to be transferred to the molten melamine in the first step. In any case, LIQUID ammonia is used to solidify the melamine once inserted into the pressure cooler 23 in Fig. 1 is gaseous or becomes gaseous instantaneously. The temperature in the pressure cooler is above 200°C and ammonia is gaseous at this temperature. There is no cooling of molten melamine with liquid ammonia. The LIQUID ammonia only exists in the pipe leading into the pressure cooler. Once in the pressure cooler, liquid ammonia flashes off.

Therefore, the large cooling step has to be achieved by a GAS to a LIQUID heat transfer, which is an order of magnitude less efficient than a LIQUID to LIQUID heat transfer. In the present invention, there might also be some cooling with gaseous ammonia but only to

temperatures above the melting point, i.e. the amount of heat removed is MUCH less than that used by Kokubo to solidify melamine. The BIC cooling step in the present invention is achieved by a LIQUID medium used to effect PHASE TRANSITION from melt to solid (slurry).

## 2. Cooling is Less Complicated and Requires Less Space

The gaseous cooling apparatus of Kokubo is large, complex and costly (see Fig. 1).

The instant process, in contrast, does not require a large, complex apparatus since the small amount of gaseous ammonia which might be used in the first cooling step of the instant process (e.g. 370°C down to e.g. 320°C) takes place at high pressure i.e. less room is required.

The quenching step, i.e. the second step in the instant process, is achieved by using an ammonia/water mixture which is always liquid throughout and therefore less space is needed.

## 3. No Ammonia Recovery Necessary

Kokubo uses water-free ammonia to solidify molten melamine in the first cooling step (cooler 23 in Fig. 1; 370°C to 270°C - 200°C).

Kokubo uses water in the second cooling step (section 18 in Fig. 1) of Kokubo.

The water ammonia mixture in cooler 2 in Fig. 1 has to be separated: first it passes through a degassification column (column 5 in Fig. 1, col. 5, line 10 of Kokubo). The gas overhead containing water and ammonia is fed into the ammonia distillation column 20. The ammonia goes overhead. In fig. 1 it is compressed and then fed back to the pressure cooler.

Such costly ammonia recovery is not necessary in the present process. The cooling medium in the present invention has NOT to be water-free. The water/ammonia mixture can be used directly for cooling WITHOUT separating the two (see page 7, lines 10-13 of the application). This saves on apparatus costs and energy costs.

The foregoing demonstrates that the instant process has considerable advantage over Kokubo. It is not a matter of yield or purity alone - the process itself is better.

She further declares that all statements made herein of her own knowledge are true, and that all statements on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States

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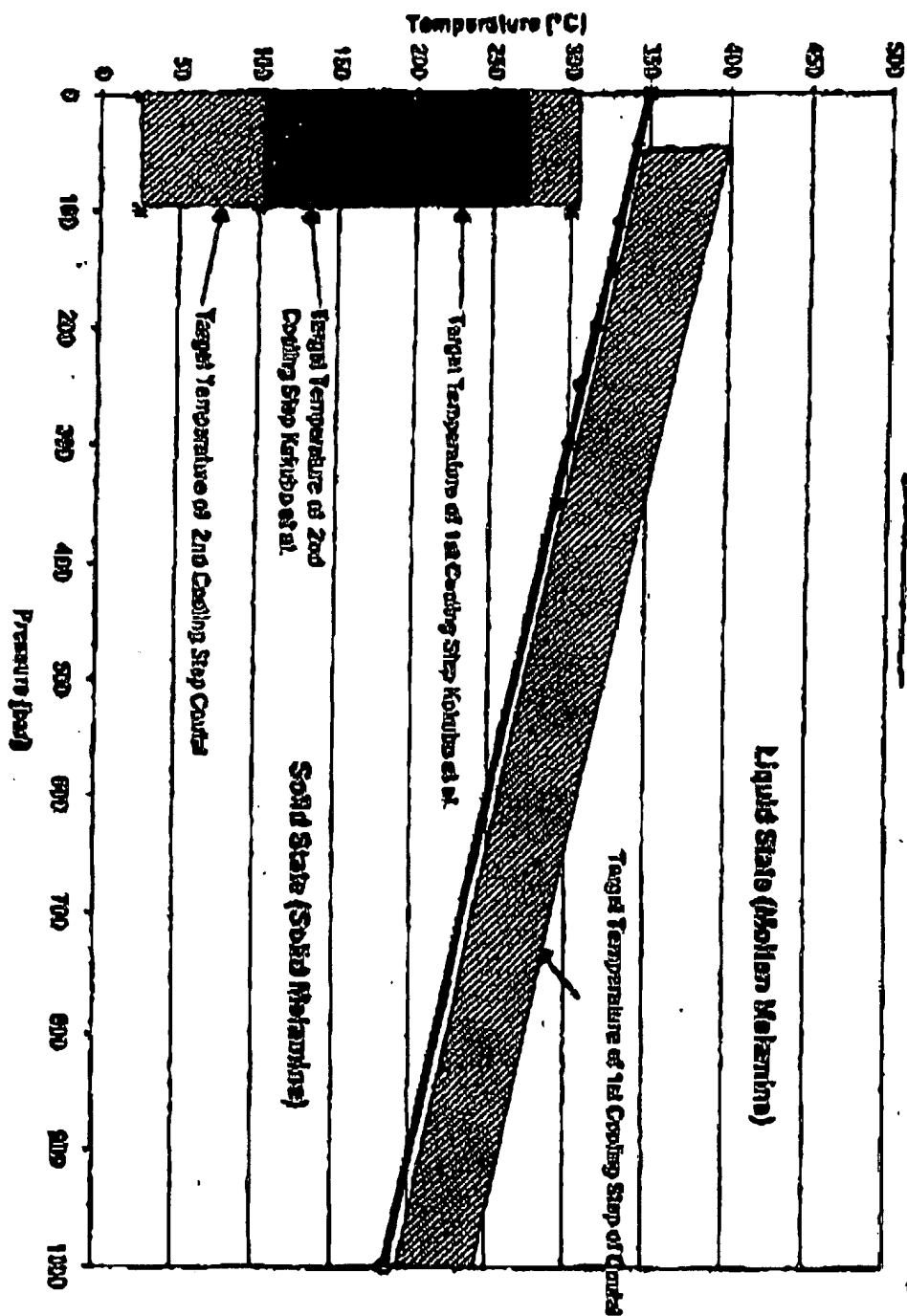
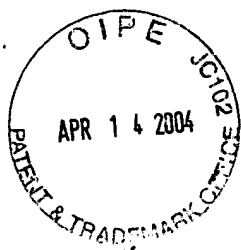
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Code, and that such willful false statements may jeopardize the validity of the application or any patent issuing thereon.

13.04.2004  
Date

Birgit Bogner  
Birgit BOGNER



Melamine Temperature in dependence of pressure  
Physical Property Source: US 6 200 385 B1 Canz et al. (Column 5, lines 13-25; Table 1 and Fig. 1)